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Influence of Containment on the Growth of Germanium-Silicon in Microgravity

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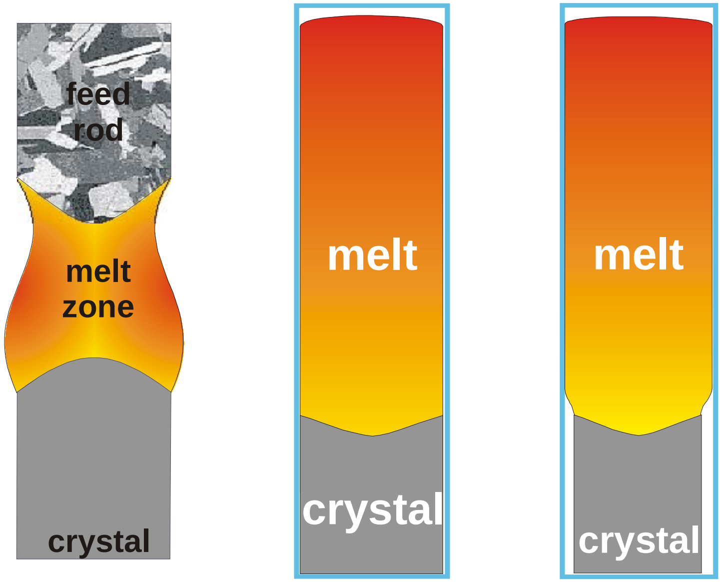
Overview of the Investigation

This investigation involves the comparison of results achieved from three types of crystal growth of germanium and germanium-silicon alloys:

- Float zone growth
- Bridgman growth
- Detached Bridgman growth

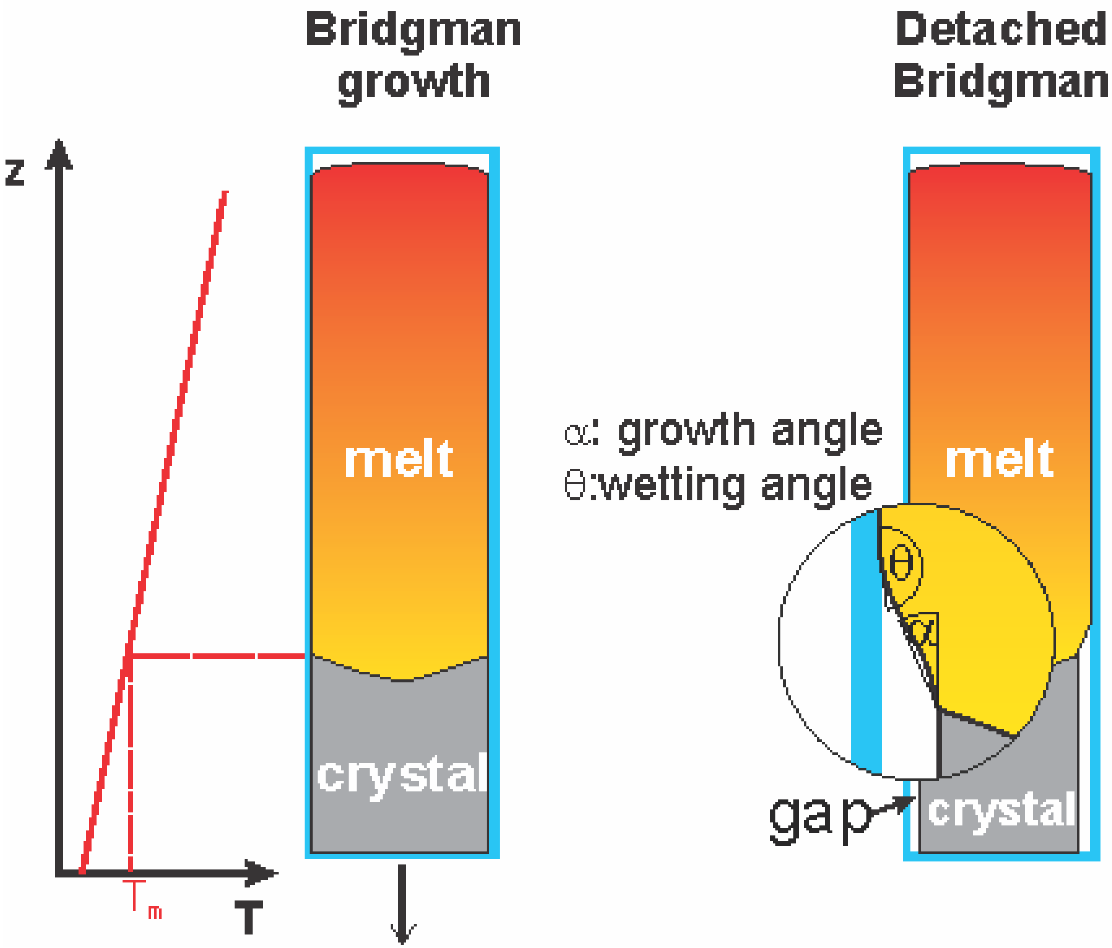


The fundamental goal of the proposed research is to determine the influence of containment on the processing-induced defects and impurity incorporation in germanium-silicon (GeSi) crystals (silicon concentration in the solid up to 5 at%) for three different growth configurations in order to quantitatively assess the improvements of crystal quality possible by detached growth.

What is Detached Bridgman Growth?

Sufficient condition for detachment[1,2]:

$$(\alpha+\theta \geq 180°)$$



Advantages

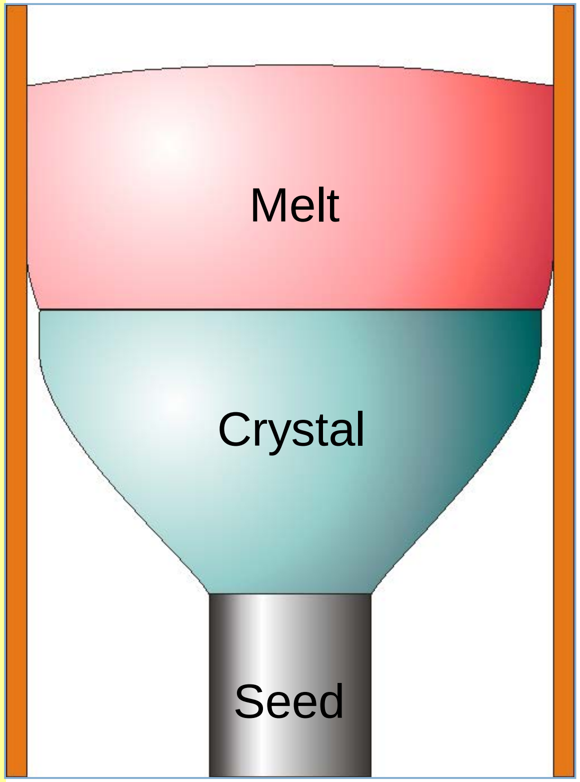
- No sticking of the crystal to the ampoule wall
- Reduced stress
- Reduced dislocations
- No heterogeneous nucleation by the ampoule
- Reduced contamination

[1] V. S. Zemskov: Fiz. Khim. Obrab. Mater. 17 (1983) 56

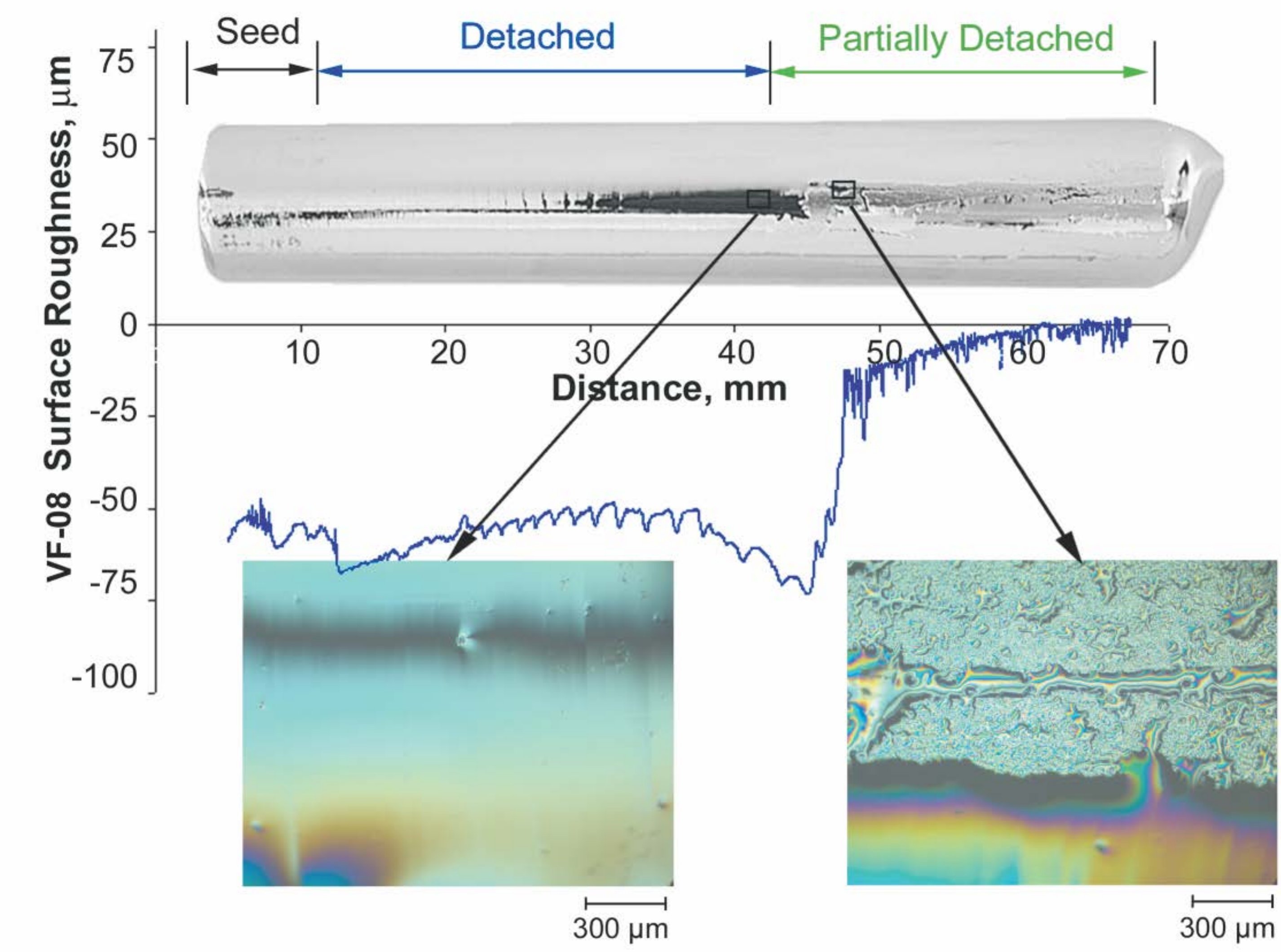
[2] T.Duffar, I.Paret-Harter, P.Dusserre: J.Crystal Growth 100 (1990) 171.

Research Motivation

- What are the conditions for detachment in microgravity and how do they depend on the governing parameters?
  - Growth angle
  - Contact angle
  - Pressure differential
  - Bond number (ratio of gravity to capillarity)
- Which detached growth solutions are dynamically stable?
- How does an initial crystal radius evolve to one of the following states?
  - Stable detached gap
  - Attachment to the crucible wall
  - Meniscus collapse
- What are the effects of angular dependence on the crystal shape (faceting effects)?



Detached Crystal Growth



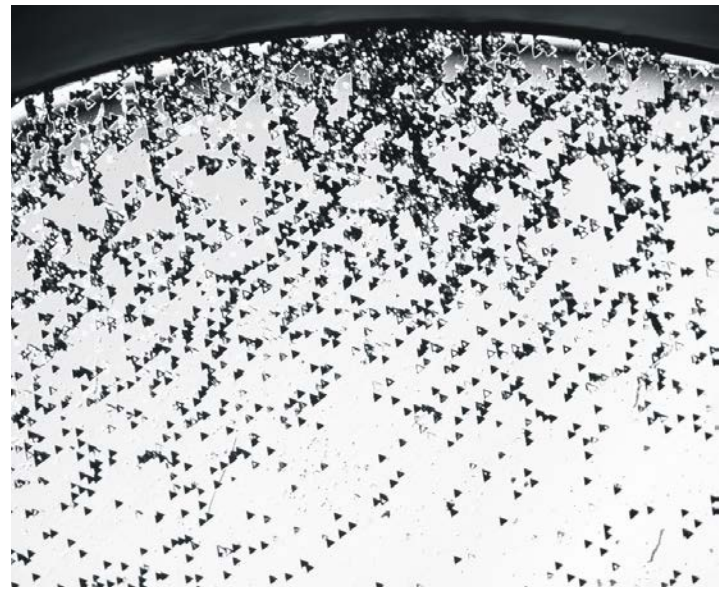
Etch Pit Densities in Detached/Attached Crystals

1mm

Completely detached grown crystal UMC7

EPD ≈ 200cm^{-2}

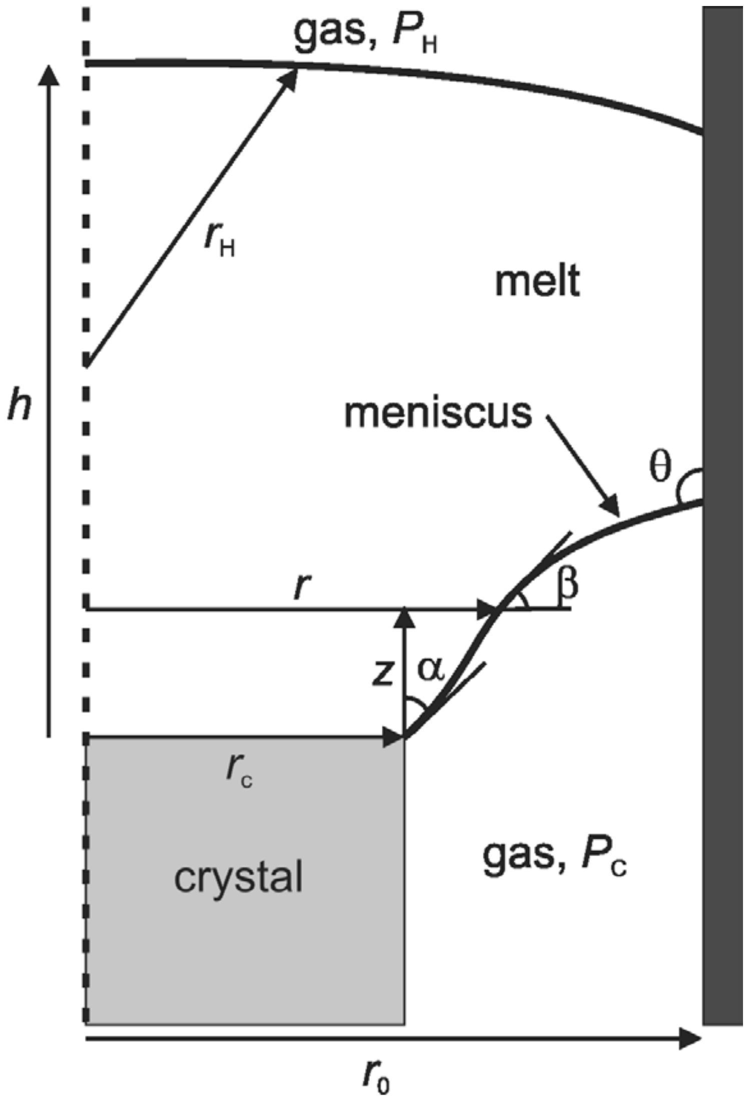
Attached grown crystal UMC6



EPD ≈ 2 ·10^{4}cm^{-2}

M. Schweizer, S. D. Cobb, M. P. Volz, J. Szoke, F. R. Szofran, JCG 235 (2002) 161-166

Schematic Diagram of Detached Solidification



α: growth angle
θ: contact or wetting angle

M. P. Volz, K. Mazuruk, *Journal of Crystal Growth* 321 (2011) 29-35

Calculation of Meniscus Shapes

$$\frac{\dfrac{d^2z}{dr^2}}{\left(1+\left(\dfrac{dz}{dr}\right)^2\right)^{3/2}}+\frac{\dfrac{dz}{dr}}{r\left(1+\left(\dfrac{dz}{dr}\right)^2\right)^{1/2}}=\Delta P-Bz(r)$$

Young-Laplace Equation

$$\Delta P=\frac{\Delta P_m r_0}{\sigma},\qquad \Delta P_m=P_H-P_C+\rho g h+2\frac{\sigma}{r_H}$$

ΔP : Dimensionless pressure differential across the meniscus

$$B=\frac{\rho g_0 r_0^{\,2}}{\sigma}$$

B = 3.248; Ge, r_0 = 6 mm
B = 4.651; InSb, r_0 = 5.5 mm

B : Bond number; ratio of gravity force to surface tension force

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$$\frac{\partial r}{\partial s}=\cos\beta,\qquad \frac{\partial z}{\partial s}=\sin\beta,\qquad \frac{\partial\beta}{\partial s}=-\frac{\sin\beta}{r}+\Delta P-Bz$$

Set of 3 coupled differential equations

Boundary Conditions

$z(0) = 0$; $\beta(0) = 90° - \alpha$;

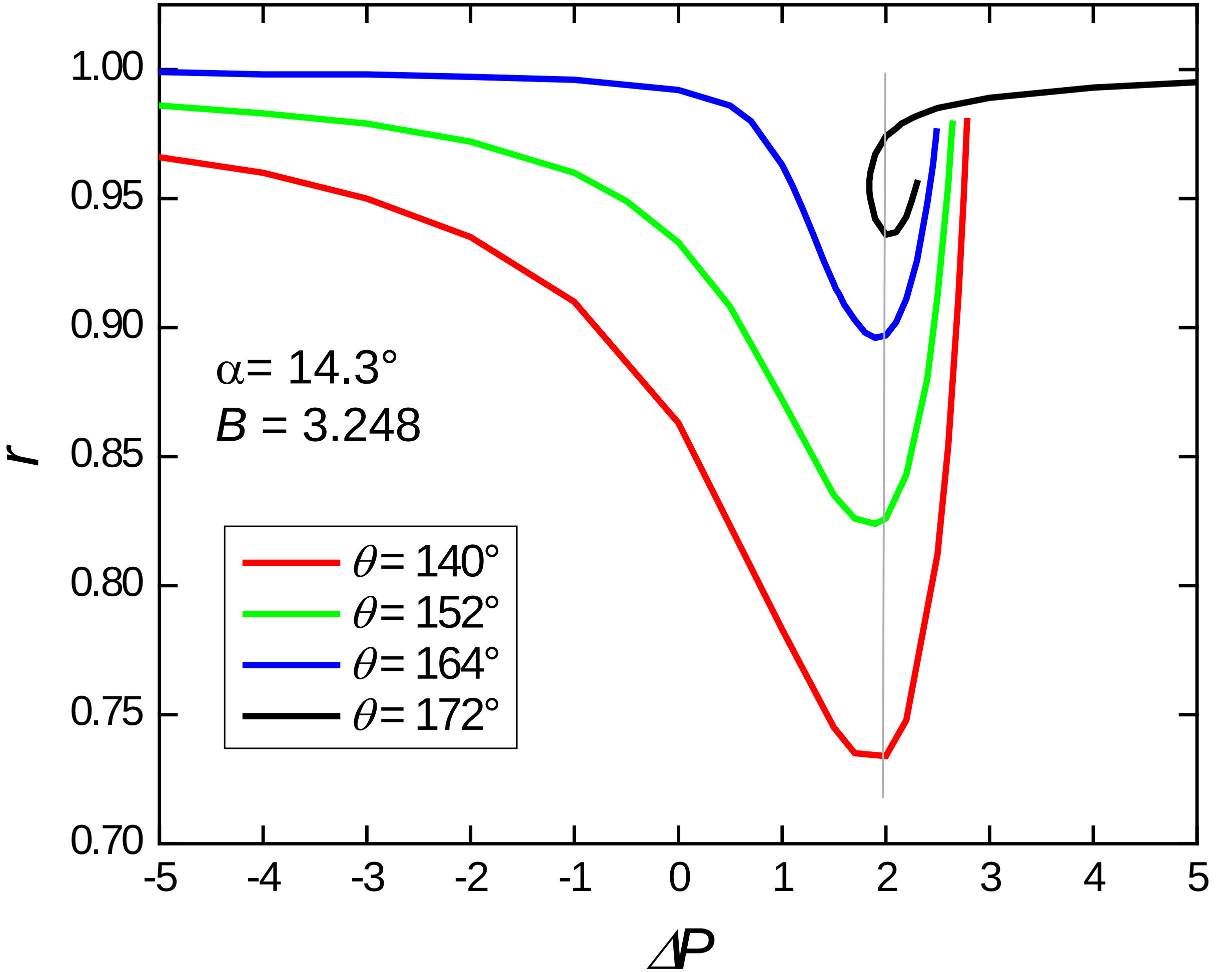
$\beta(1) = \theta - 90°$; $r(1) = 1$

α: growth angle
θ: contact or wetting angle

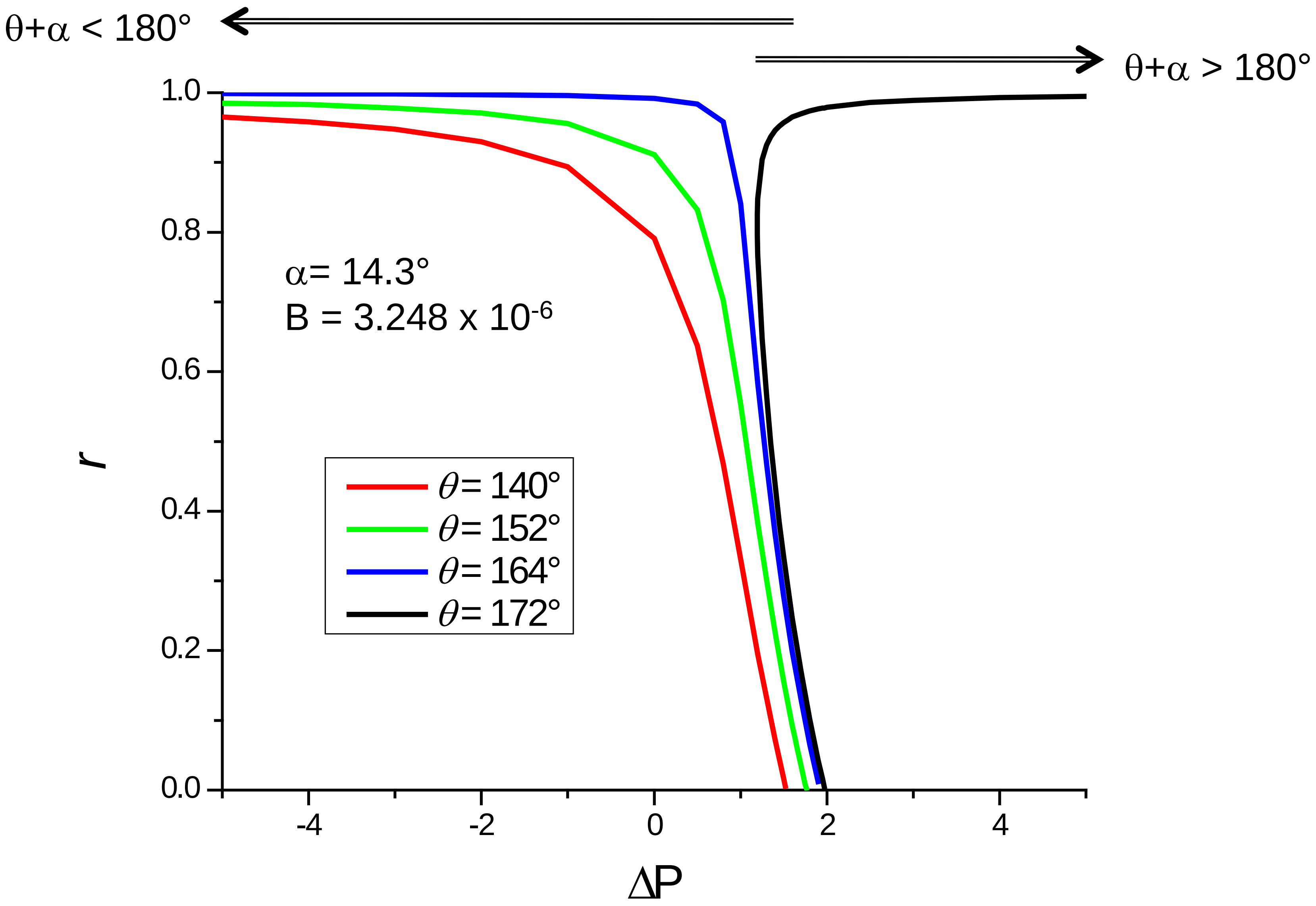
Gap Width vs. Pressure Differential (Ge at 1g)

$\theta+\alpha < 180°$

$\theta+\alpha > 180°$

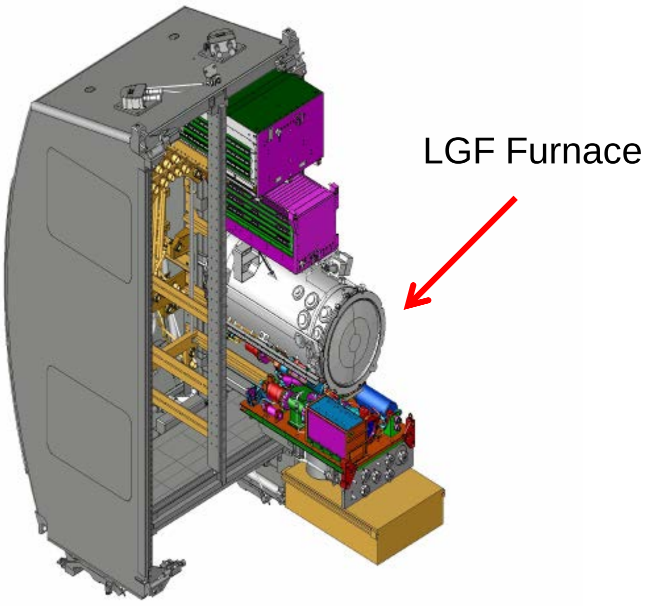


Gap Width vs. Pressure Differential (Ge at 10^{-6} x g_0)



ICESAGE Flight Investigation

- “Influence of Containment on the Growth of Silicon-Germanium” (ICESAGE) is a collaborative investigation between NASA and the European Space Agency (ESA)
- The ICESAGE experiments will be conducted in the Low Gradient Furnace (LGF) in the Materials Science Laboratory on the International Space Station (ISS)
- Processing parameters will be varied to asses their affect on detachment
  - Sample Material (GeSi, Ge:Ga)
    - Affects the growth angle
    - Comparison of semiconductor alloy and doped element
  - Inner Ampoule surface material (SiO_2, boron nitride)
    - Affects the contact angle
  - Pressure: positive, negative, or zero (vacuum) gas pressure below the meniscus



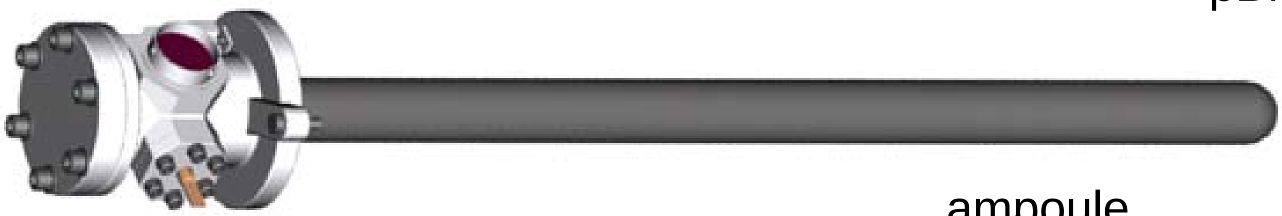
Materials Science Laboratory

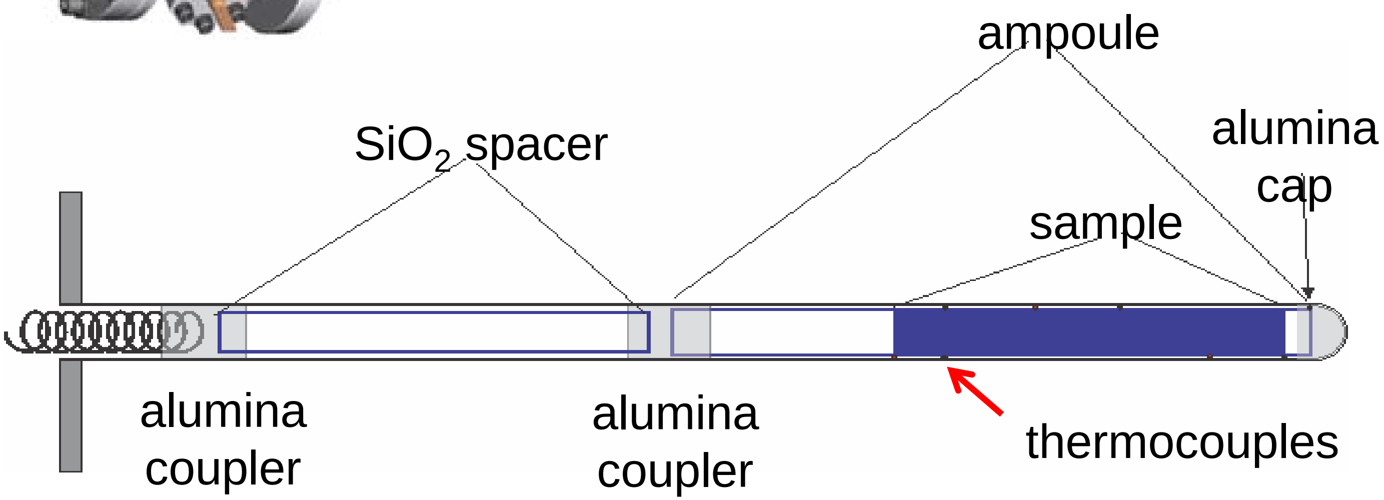
Microgravity Effects

- Microgravity reduces the pressure head (ρgh) resulting from the weight of the melt.
  - Detached growth requires that capillary forces dominate over gravitational forces.
  - On Earth, gravity complicates a comparison of detached growth theory and experiment: the pressure head continuously decreases as the melt solidifies and the pressure varies along the height of the meniscus.
- Microgravity allows a larger value of the gap width.
  - On Earth, when the gap width becomes too large, gravity overcomes surface tension, a stable meniscus cannot be maintained, and the melt will flow down between the crystal and ampoule wall.
  - A large initial gap width will allow measurement of anisotropy in the growth angle.
- Microgravity enables a study of the dynamic stability of crystallization independent of thermal effects.

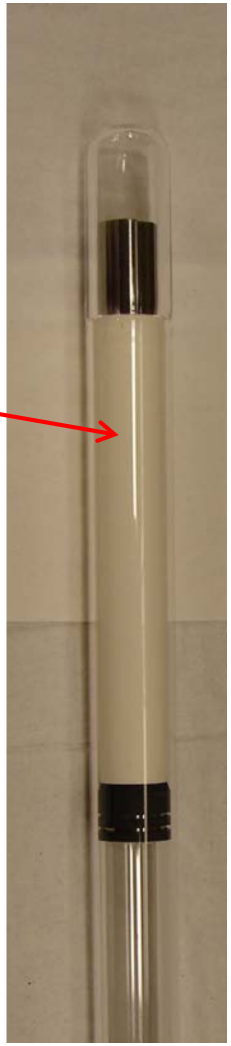
Ampoule and Cartridge Layout

- A $Ge_{1-x}Si_x$ ingot is placed inside a pyrolitic boron nitride (pBN) tube and sealed in a SiO_2 ampoule.
- The ampoule is placed inside a cartridge which is inserted into the furnace.
- Thermocouples in the cartridge provide for real-time monitoring of the thermal profile

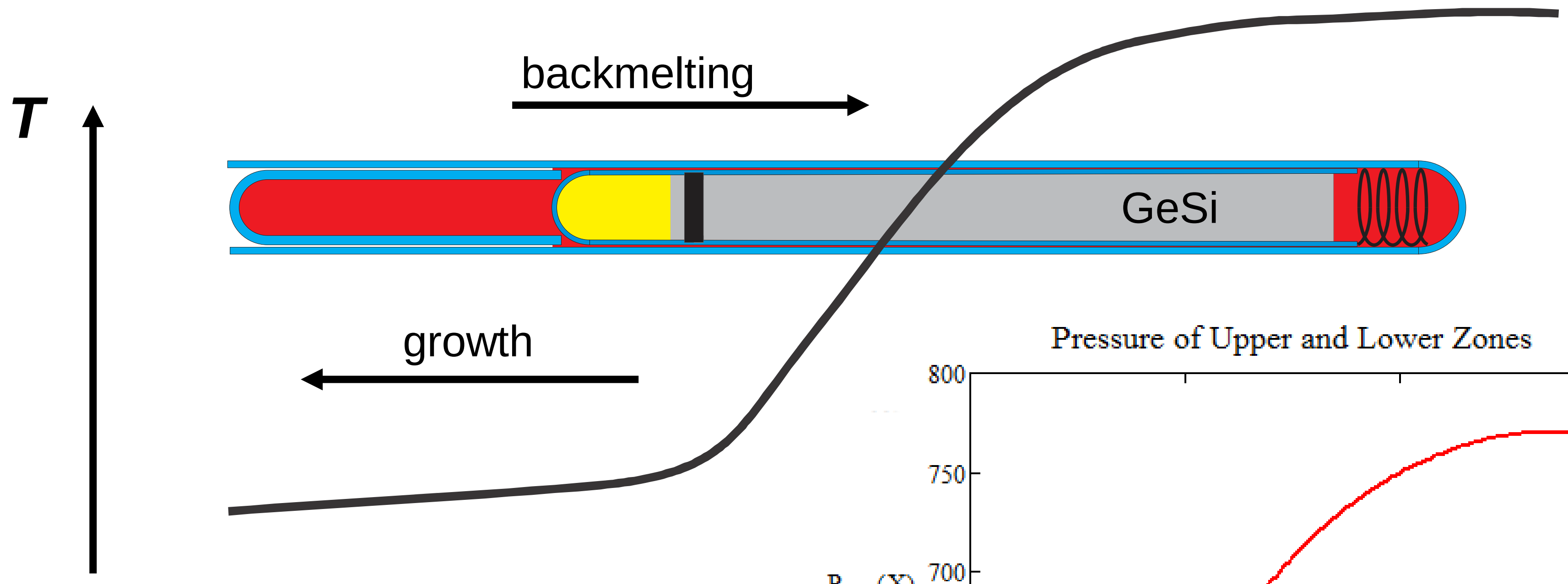


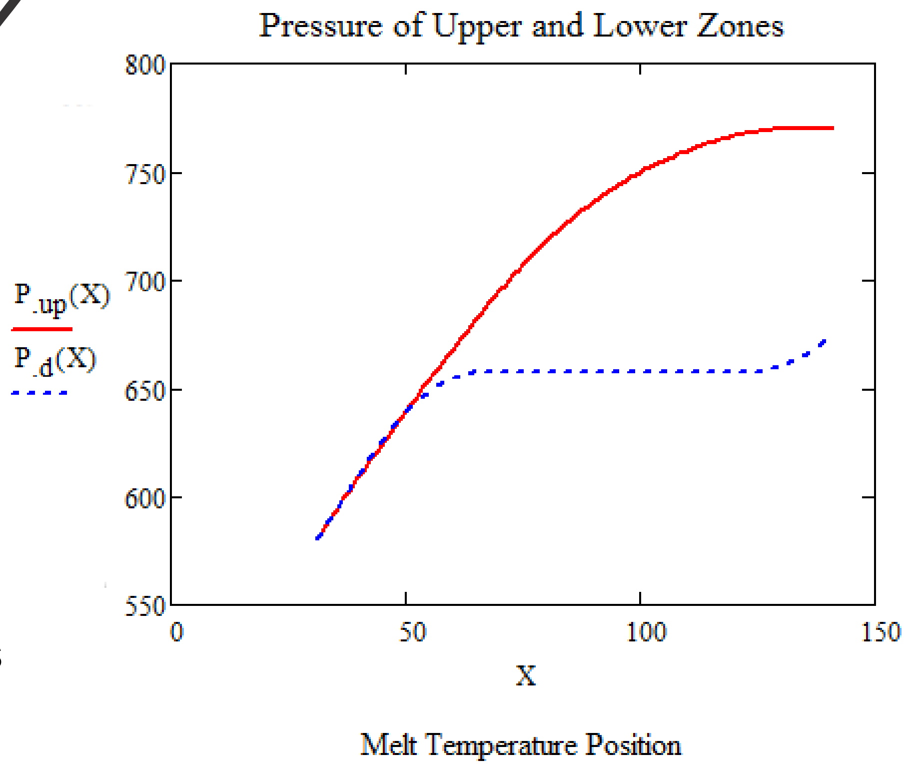


pBN tube



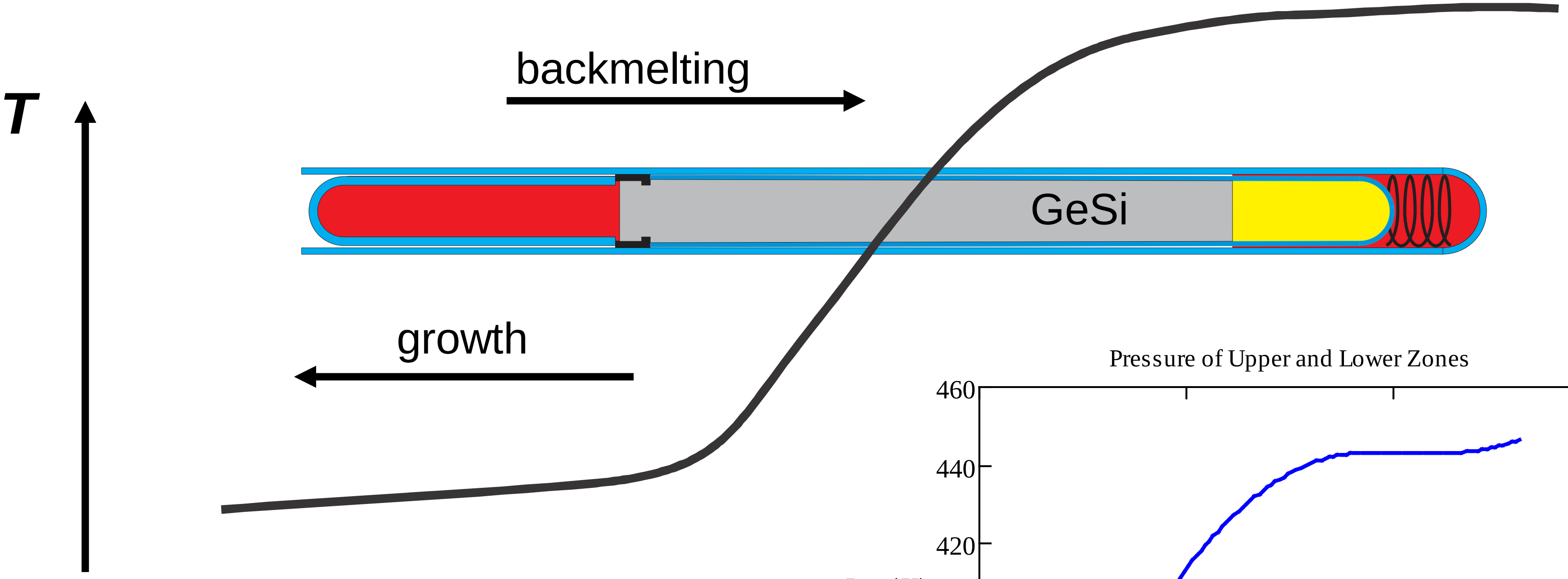
Positive Pressure Configuration

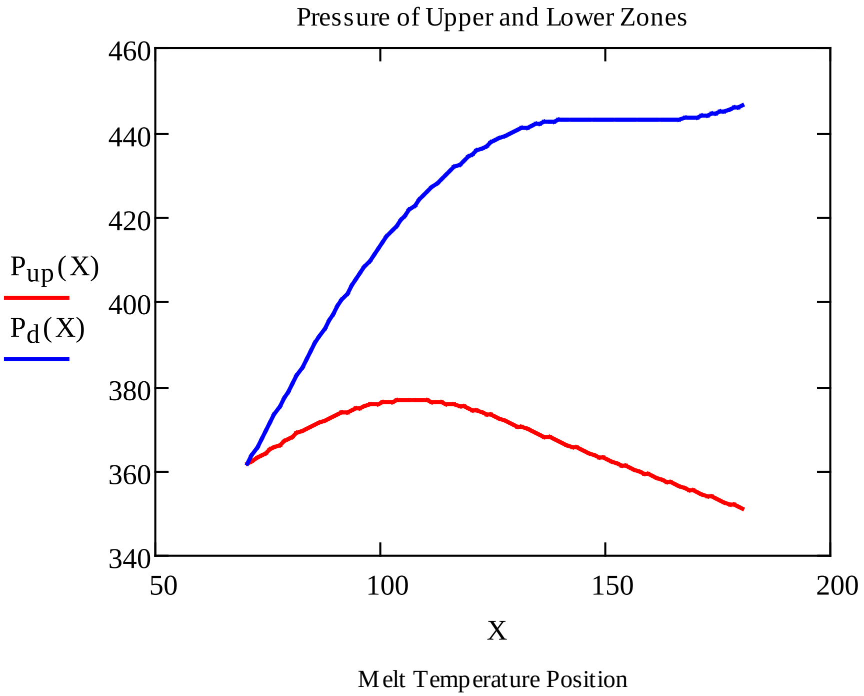




- When the top of the ingot melts, different pressure volumes are created
- As the ingot is inserted into the furnace, the pressure above the meniscus increases faster than below the meniscus

Negative Pressure Configuration





- When the top of the ingot melts, different pressure volumes are created
- As the ingot is inserted into the furnace, the pressure below the meniscus increases faster than above the meniscus

Summary

- Crystals grown by the detached Bridgman method have greatly increased crystalline perfection, motivating a systematic study of the phenomenon
- A theory describing the conditions for detachment has been developed
- Only crystals where $\alpha + \theta > 180°$ are expected to achieve stable detached growth in microgravity
- Reproducible detached growth has been achieved in the laboratory under limited conditions
- Microgravity will allow the study of detachment over a range of parameters not possible to achieve on Earth
- A series of Ge and GeSi crystal growth experiments are being developed for processing on the ISS